

# **Technical Report**

on

**Title of proposed project:**

**“Exploration of Advanced Ceramic Gain Media with Broadband for the  
application to High Energy Laser”**

**AOARD Grant Number:FA2386-11-1-4007**

by

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**Submission Date: August 1<sup>st</sup>, 2013**

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Report Documentation Page		Form Approved OMB No. 0704-0188
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1. REPORT DATE <b>15 AUG 2013</b>	2. REPORT TYPE <b>Final</b>	3. DATES COVERED <b>07-12-2010 to 05-07-2013</b>
4. TITLE AND SUBTITLE <b>Exploration of advanced ceramic gain media with broadband for the application to high energy laser.</b>		5a. CONTRACT NUMBER <b>FA23861114007</b>
		5b. GRANT NUMBER
		5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S) <b>Tomosumi Kamimura</b>		5d. PROJECT NUMBER
		5e. TASK NUMBER
		5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) <b>Osaka Institute of Technology,5-16-1 Ohmiya, Asahi-ku,Osaka 535-8585,Japan,NA,NA</b>		8. PERFORMING ORGANIZATION REPORT NUMBER <b>N/A</b>
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) <b>AOARD, UNIT 45002, APO, AP, 96338-5002</b>		10. SPONSOR/MONITOR'S ACRONYM(S) <b>AOARD</b>
		11. SPONSOR/MONITOR'S REPORT NUMBER(S) <b>AOARD-114007</b>
12. DISTRIBUTION/AVAILABILITY STATEMENT <b>Approved for public release; distribution unlimited</b>		
13. SUPPLEMENTARY NOTES		
14. ABSTRACT <b>Practical laser generation from polycrystalline ceramic materials has become available; it is expected that ceramic laser technology will completely surpass conventional glass and single crystal laser technologies in the future. In this study, the authors? fabricated advanced ceramic gain media such as CLNGG (Ca3LixNb1.5-xGa(3.5-2x)O12), CLNTGG (Ca3(LiNbTaGa)5O12)) and YSAG (Y3Sc1Al4O12), and investigated spectroscopic properties including broadband emission spectrum and fluorescence lifetime. The best candidate compositions were screened for high energy lasing and fabricated into high optical quality ceramic gain media for examination using continuous wave-generation laser oscillation tests. Nd doped Y3ScxAI5-xO12 (x = 0 to 2) ceramics were confirmed as one of the candidates for high energy laser applications. For the first time, a composite structure with three Nd doped Y3ScxAI5-xO12 (x = 0 to 2) ceramic compositions was successfully fabricated to obtain a broadband spectrum. The emission spectrum of multi-layered YSAG composite was more than 5 times broader compared to that of 1at% Nd:YAG. The laser oscillation test showed that multi-layered composite made of Nd:YSAG ceramics reached laser grade optical quality.</b>		
15. SUBJECT TERMS <b>High Energy Lasers, Transparent Ceramics, Transparent Polycrystalline Materials, Advanced Ceramic Gain Media, Broadband, YSAG (Y3Sc1Al4O12)</b>		

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT <b>Same as Report (SAR)</b>	18. NUMBER OF PAGES <b>17</b>	19a. NAME OF RESPONSIBLE PERSON
a. REPORT <b>unclassified</b>	b. ABSTRACT <b>unclassified</b>	c. THIS PAGE <b>unclassified</b>			

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Acknowledgements

## **Title of Project: “Exploration of Advanced Ceramic Gain Media with Broadband for the application to High Energy Laser”**

### **Abstract**

Practical laser generation from polycrystalline ceramic materials has become available; furthermore, it is expected that ceramic laser technology will completely surpass the conventional glass and single crystal laser technologies in the future. In this work, we fabricated advanced ceramic gain media for broadband such as CLNGG ( $\text{Ca}_3\text{Li}_x\text{Nb}_{1.5-x}\text{Ga}_{(3.5-2x)}\text{O}_{12}$ ), CLNTGG ( $\text{Ca}_3(\text{LiNbTaGa})_5\text{O}_{12}$ ) and YSAG ( $\text{Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}$ ), and investigated their spectroscopic properties such as emission spectrum and fluorescence lifetime. Among them, we screened the best candidate compositions for high energy laser and developed into high optical quality ceramic gain media. Nd doped  $\text{Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ( $x = 0$  to 2) ceramics was confirmed as one of the candidates for high energy laser with broadband. A composite structure with three compositions in Nd doped  $\text{Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ( $x = 0$  to 2) ceramics was successfully fabricated for the first time to obtain a broadband spectrum. The emission spectrum of multi-layered YSAG composite was more than 5 times broader compared to that of 1at% Nd:YAG. The laser oscillation test showed that multi-layered composite made of Nd:YSAG ceramics reached laser grade optical quality. Based upon the results of this work, we will apply advanced ceramic materials for high energy laser application in the future.

Keywords: Transparent ceramics, transparent polycrystalline material, high energy laser, advanced ceramic gain media, broadband, YSAG ( $\text{Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}$ )

### **1. Introduction**

Recently high performance polycrystalline ceramic laser, which can exceed the performance of single crystal in major solid-state lasers, has been developed. The wide range of materials which can be prepared in optically transparent polycrystalline form has opened the door to the development of many new laser host/dopant combinations which may have properties such as spectral line width, wavelength, quantum efficiency, and thermo-mechanical properties that are not obtainable from single crystal hosts. Since ceramic materials have more advantages than single crystal and glass laser materials, innovative ideas and their applications are expected in high energy laser research.

Basic properties needed for laser material are large product of  $\sigma$  (cross-section of stimulated emission) and  $\tau$  (life time of fluorescence), high thermal conductivity for material cooling and chemical stability. Especially, broadening for absorption and emission spectrum are important in the high energy laser field. Broadening of the spectrum in the laser material gives functionality such as tunability and short pulse lasing of the wavelength. In contrast, in

broadening of the absorption, the temperature adjusting of the pumping LD becomes unnecessary. And also, flexible scale-up is demanded to the dimension of the laser material for industrial practical use. However, conventional high energy laser material such as glass and single crystal materials do not satisfy these requirements. The glass material has extremely low laser repetition rate due to its low thermal conductivity (just about one-tenth (1/10) of single crystal materials). Power scaling (TW or PW) is impossible because it is difficult to grow a large size single crystal laser material. A promising approach which can meet the requirements for high energy field is the development of advanced ceramic laser gain media with broadband because ceramic materials already have good thermo-mechanical properties, and it is possible to produce large scaled gain media by ceramic manufacturing process.

## **2. Objective of the project**

The objective of the project is to explore the promising materials with broadband and investigation of their basic spectrum properties for high energy laser. Material team will develop advanced garnet ceramics having the complicated composition, namely CLNGG ( $\text{Ca}_3\text{Li}_x\text{Nb}_{1.5-x}\text{Ga}_{(3.5-2x)}\text{O}_{12}$ ), CLNTGG ( $\text{Ca}_3(\text{LiNbTaGa})_5\text{O}_{12}$ ) and YSAG ( $\text{Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}$ ). Evaluation team will investigate their spectroscopic properties such as emission spectrum and lifetime. We will screen the best candidate compositions for high energy laser and fabricate into high optical quality ceramic gain media. In addition, possibility of new technology applications such as composite structure will be challenged to develop an advanced garnet ceramics having the complicated structure. If good sample is achieved, laser oscillation tests for cw-generation will be performed.

## **3. Project schedule and targets**

The project will be processed on the exploration of advanced ceramic gain media for broadband, and will investigate their emission, lifetime, spectroscopic properties. Materials exploration and its evaluation will be involved; (1) advanced garnet ceramics having the complicated composition such as CLNGG, CLNTGG and YSAG, (2) investigation of technology for making transparent material in advanced garnet ceramics, (3) investigation of technology for making composite structure having complicated compositions of YSAG, (4) evaluation of their spectroscopic properties such as emission spectrum and lifetime, (5) if good optical quality sample is achieved, laser oscillation tests for cw-generation will be performed. It is planned to run as two-year research project as shown in Fig.3-1, and targets for each year term are summarized in table 3-1. In the first year, exploration of advanced ceramic gain media for broadband will be performed for advanced garnet ceramics, and spectroscopic properties such as emission spectrum and lifetime will be evaluated to

screening an optimum complicated composition.

	First Year					Second Year				
	2011 Q1	2011 Q2	2011 Q3	2011 Q4	2012 Q1	2012 Q2	2012 Q3	2012 Q4	2013 Q1	2013 Q2
Fabrication of Material	(1) Exploration of advanced garnet ceramics (CLNGG, CLNTGG, YSAG)									
						(3) Investigation of technology for making transparent material				
						(2) Investigation of technology for making composite structure having complicated compositions of YSAG				
Evaluation of Material										
	(4) Evaluation of investigate their spectroscopic properties such as emission spectrum and lifetime									
						(5) If good sample is achieved, laser oscillation tests for cw-generation will be performed.				

Fig.3-1 Schedule of this project.

Table 3-1 Targets for each year term of the project

	Target for each year
First year (2011)	Ceramic material development for high energy lasers
Second Year (2012)	Material evaluation and laser testing

#### 4. Fabrication of advanced garnet ceramics

##### 4.1 CLNGG and CLNTGG ceramics

Fabrication process for advanced garnet ceramics is shown in figure 4-1. High purity (>99.99%) powders,  $\text{CaCO}_3$ ,  $\text{Li}_2\text{CO}_3$ ,  $\text{Nb}_2\text{O}_5$ ,  $\text{Ta}_2\text{O}_5$ , and  $\text{Ga}_2\text{O}_3$  were used as starting materials. For laser active dopants ( $\text{Nd}_2\text{O}_3$ ,  $\text{Yb}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$  and  $\text{Tm}_2\text{O}_3$ ), also high purity (>99.99%) powders were used. (Abbreviation for  $\text{Ca}_3\text{Li}_x\text{Nb}_{(1.5-x)}\text{Ga}_{(3.5-2x)}\text{O}_{12}$  and  $\text{Ca}_3(\text{LiNbTaGa})_5\text{O}_{12}$  is written as CLNGG and CLNTGG, respectively.) These powders were weighted to have desired garnet compositions (i.e., CLNGG or CLNTGG), and they were mixed by ball milling process in alcohol solvent with sintering aids and a little amount of binder for tablet formation. The milled slurry was dried to obtain powders. Small tablets were prepared by using a metal mold (diameter: 12mm) and CIP machine (isostatic pressure:

200MPa). Then the tablets were sintered under oxygen atmosphere around 1400°C for about 5 hours. Translucent ceramics were obtained after sintering. Then the sintered samples were polished to evaluate their properties and some samples were thermally etched to inspect the microstructure. Appearance of polished CLNGG and CLNTGG ceramic samples are shown in figure 4-2.

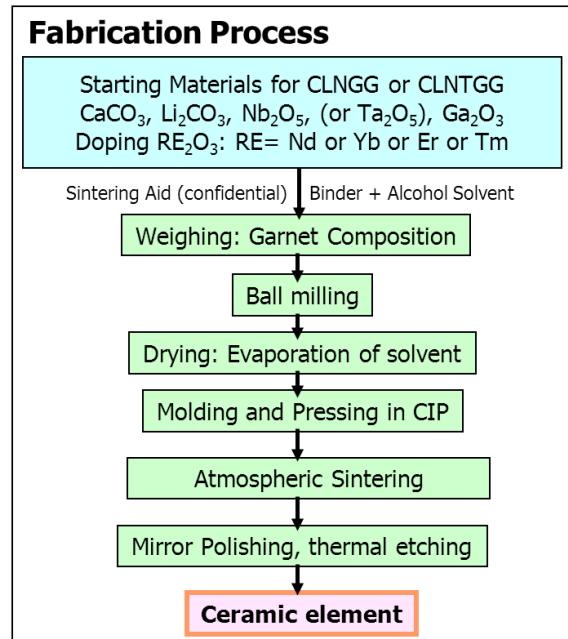


Figure 4-1 Flow-chart for fabrication of CLNGG and CLNTGG ceramics.

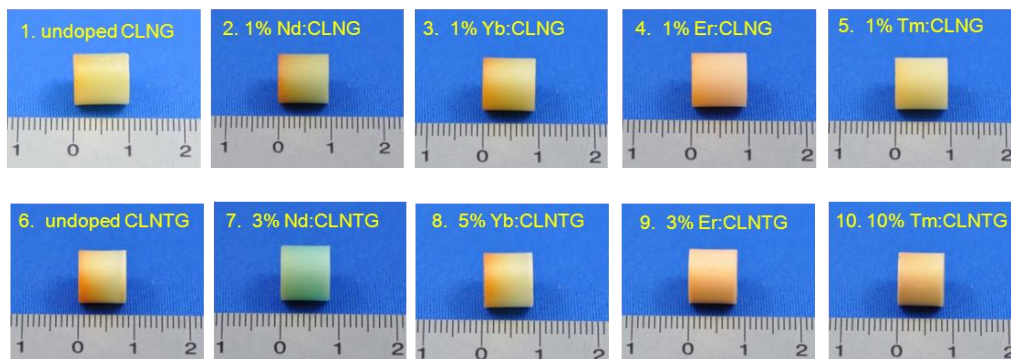


Figure 4-2 Appearance of a series of CLNGG and CLNTGG ceramics

#### 4.2 Nd:YSAG Ceramics

Fabrication process for Nd:YSAG ceramic is shown in figure 4-3. High purity (>99.99%) powders, Nd<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, Sc<sub>2</sub>O<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub> were used as starting materials. These powders were weighed to have garnet composition of Nd doped Y<sub>3</sub>Sc<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub> (x = 0 to 2), and they were mixed by ball milling process in alcohol solvent with sintering aids and a little amount of

binder. The milled slurry was dried to obtain powders. Small tablets were prepared by using a metal mold (diameter: 12mm) and CIP machine (isostatic pressure: 200MPa). After removing binder, the tablets were sintered in vacuum around 1750°C for about 10 hours. Transparent ceramics were obtained after vacuum sintering. Then the sintered samples were polished to evaluate their properties and some samples were thermally etched to inspect the microstructure. Appearance of polished Nd:YSAG ceramic samples are shown in figure 4-4.

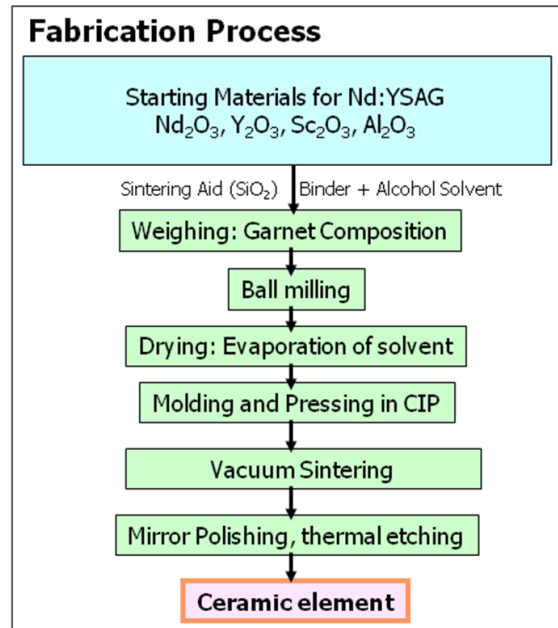


Figure 4-3 Flow-chart for fabrication of Nd:YSAG ceramics.

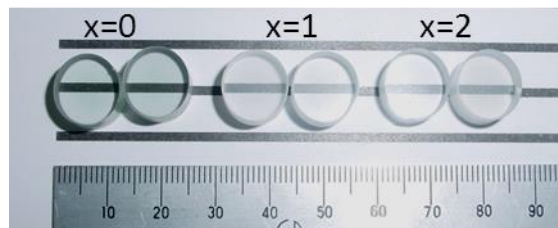


Figure 4-4 Appearance of polished Nd:Y<sub>3</sub>Sc<sub>x</sub>Al<sub>5-x</sub>O<sub>12</sub> ceramics

## 5. Observation on microstructure of advanced garnet ceramics

Microstructures of CLNGG, CLNTGG and Nd:YSAG ceramics were shown in figure 5-1, 5-2 and 5-3, respectively. As seen in their microstructures, residual pores were hardly observed on the surface of the specimens, suggesting that the materials are almost fully densified after sintering. Also, no secondary phases or segregation were confirmed under an optical polarized microscope, suggesting that garnet single phase was formed.

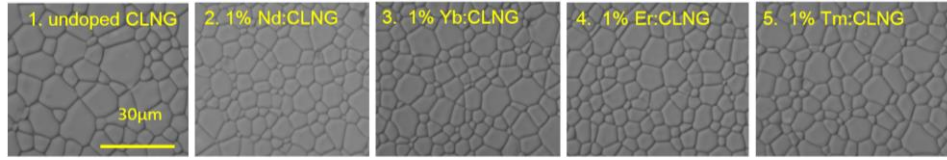


Figure 5-1 Microstructure of CLNGG ceramics.

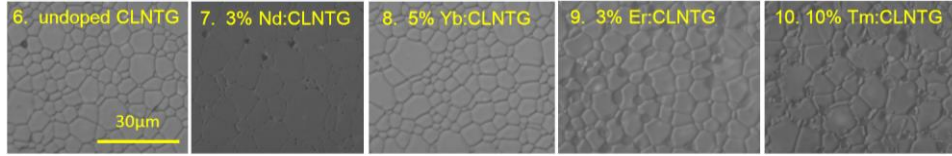


Figure 5-2 Microstructure of CLNTGG ceramics.

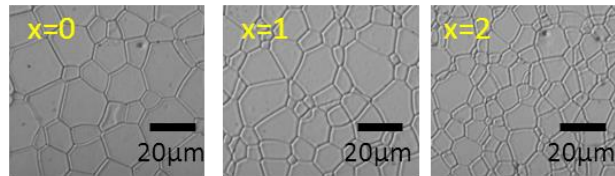


Figure 5-3 Microstructure of  $\text{Nd:Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ceramics.

## 6. Evaluation of spectroscopic properties

### 6.1 CLNGG and CLNTGG ceramics

The measurement system of the photo luminescence (PL) of the ceramic sample is shown in figure 6-1. A chopped pumping light was irradiated to the surface of prepared ceramic sample with a focusing lens. In the case of  $\text{Nd}^{3+}$  ion, an LD (CW) of 808nm was used as a pump light source. In addition, 513.5nm was used for  $\text{Er}^{3+}$  ion, 946nm was used for  $\text{Yb}^{3+}$  ion, and a 785nm was used for  $\text{Tm}^{3+}$  ion, respectively.

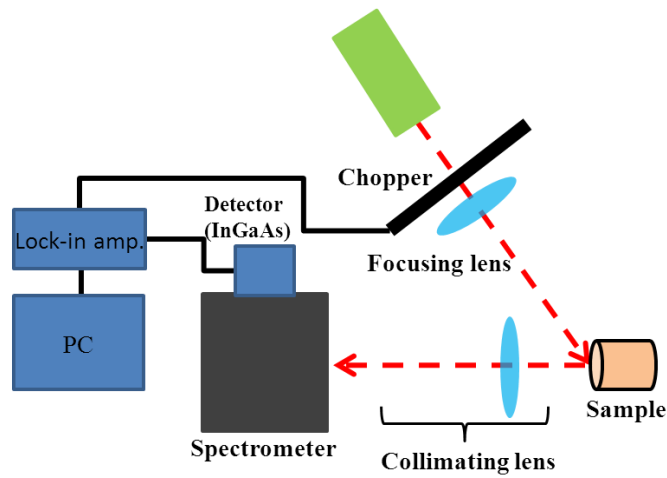


Figure 6-1 Measurement system of the photo luminescence of the ceramic sample.

The emission from a ceramic sample was collimated to obtain an optimum intensity at the spectrometer. Spectrum of emission was detected with a spectrometer and an InGaAs detector controlled in PC and lock-in amplifier. Since the measurement was conducted at room temperature (300K), the spectra were not sharp.

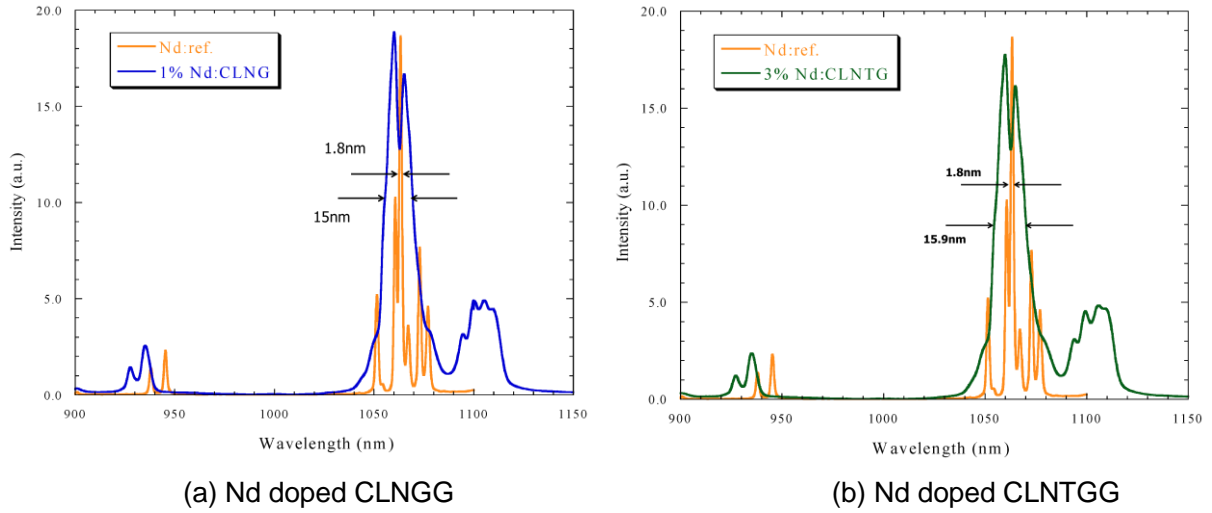


Figure 6-2 Measurement results of emission spectrum.

Measurement results of Nd doped CLNGG and CLNTGG are shown in figure 6-2(a) and (b), respectively. The emission spectrum for co-doping of Li, Nb, Ta or Ga into garnet structure was largely broadened compared to emission spectral width of Nd:YAG ceramic (1.8nm). The emission spectral width for the Nd doped CLNGG sample, was 15nm, and for Nd doped CLNTGG was 15.9nm. Crystal field for Nd was enhanced and hence the spectrum was broadened. CLNTGG has a broader spectrum than CLNGG due to its more complicated composition.

The measurement system of fluorescence lifetime is shown in figure 6-3. A pumping laser operated with pulse mode was irradiated into the ceramic sample. Fluorescence decay from the edge of the pulse was measured by using an InGaAs detector and a photon counter controlled with PC. The measurement wavelength was the peak wavelength of the emission spectrum. Measurement results of the CLNGG ceramic sample (Nd doped contents: 1at%, 3at% and 5at%) are shown in figure 6-4. Fluorescence lifetime of 1at% Nd:CLNGG ceramics was 224 $\mu$ s, which is same as in the case of 1at%Nd:YAG ceramic. The fluorescence lifetime became shorter with an increase of Nd doping amount. Fluorescence lifetime for the 3at%Nd:CLNGG ceramic sample was 165 $\mu$ s, and for the 5at%Nd:CLNGG ceramic sample was 97 $\mu$ s, respectively.

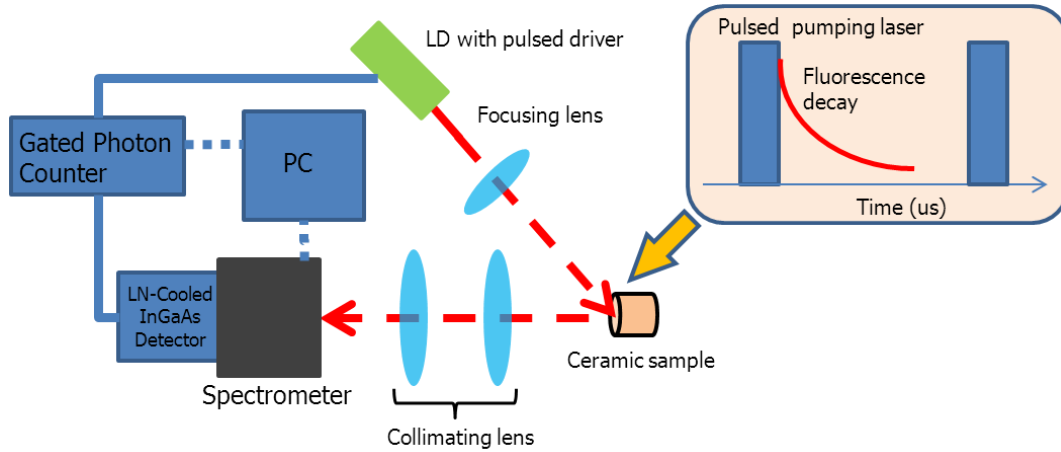


Figure 6-3 Measurement system for fluorescence lifetime.

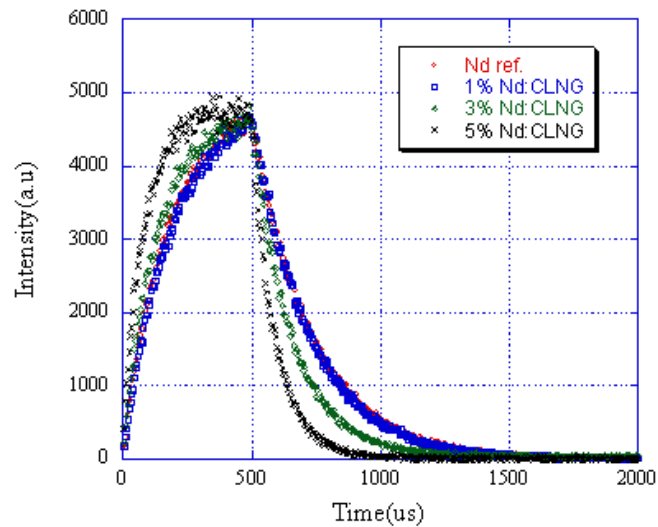


Figure 6-4 Fluorescence lifetime of the CLNGG ceramic sample.

Measurement results of emission spectral width and fluorescence lifetime of CLNGG and CLNTGG are summarized in table 6-1. Broadband emissions that cannot be observed in normal YAG or sesquioxide materials were confirmed. In the CLNGG and CLNTGG structure, the condition of distorted symmetry of crystal around the laser active ions in a lattice of ceramics was thought to make a spectrum broader, which is similar to the case of amorphous materials. However, fluorescence lifetime was similar to the existing materials. In addition, it is difficult to obtain a transparent quality with laser grade under vacuum or reducing atmosphere because gallium (Ga) and niobium (Nb) have very low affinity with oxygen. If a fabrication technology to produce laser grade can be established, it will become a promising new laser material.

Table 6-1 Measurement results of emission spectral width and fluorescence lifetime of CLNGG and CLNTGG ceramics

No.	Host Materials	Dopant (Laser active ions)	Doping Concentration (at.%)	Increased peak spectrum width (nm)	Fluorescence lifetime (us)
1	CLNGG	undoped	0%	-----	-----
2		Nd	1%	13.2	224
3		Yb	1%	11.6	695
4		Er	1%	9.7	7782
5		Tm	1%	16.2	616
6	CLNTGG	undoped	0%	-----	-----
7		Nd	3%	14.1	172
8		Yb	5%	14.3	742
9		Er	3%	15.1	8288
10		Tm	10%	15.5	992

## 6.2 Nd:YSAG Ceramics

Emission spectrum and fluorescence lifetime of the Nd:YSAG ceramic samples were measured in the same way as described above (see figure 6-1). Obtained results are summarized in figure 6-5 (a) and (b). Since the resolution of the measurement system used for this work was about 1nm, it is not possible to compare the details. As for comparison, emission spectrum of 1at%Nd:YAG that was measured with high resolution system was included as a reference in the figure 6-5(a). [1] Emission bandwidth of the 1at%Nd:YAG was 1.1nm in FWHM. Compared to this value, the emission spectra of the YSAG ceramics with complicated compositions were broaden more than 5 times at a maximum. It was confirmed that emission spectral shift and width can be controlled with a composition ratio of Sc to Al in the case of Nd doped  $Y_3Sc_xAl_{5-x}O_{12}$  ( $x = 0$  to 2). When the Sc to Al ratio become larger, the fluorescence lifetime has become longer and spectral width has become larger compared to that of 1at%Nd:YAG (reference). The fluorescence lifetime for 1at% of Nd:YSAG (Sc:Al = 1:4) increased about 10 $\mu$ s as compared with 246 $\mu$ s of the Nd:YAG. Moreover, the fluorescence lifetime of 1at% Nd:YSAG (Sc:Al = 2:3) increased to 284 $\mu$ s. From these results, Nd doped  $Y_3Sc_xAl_{5-x}O_{12}$  ( $x = 0$  to 2) ceramics can be considered as one of the candidates for high energy laser materials with broadband.

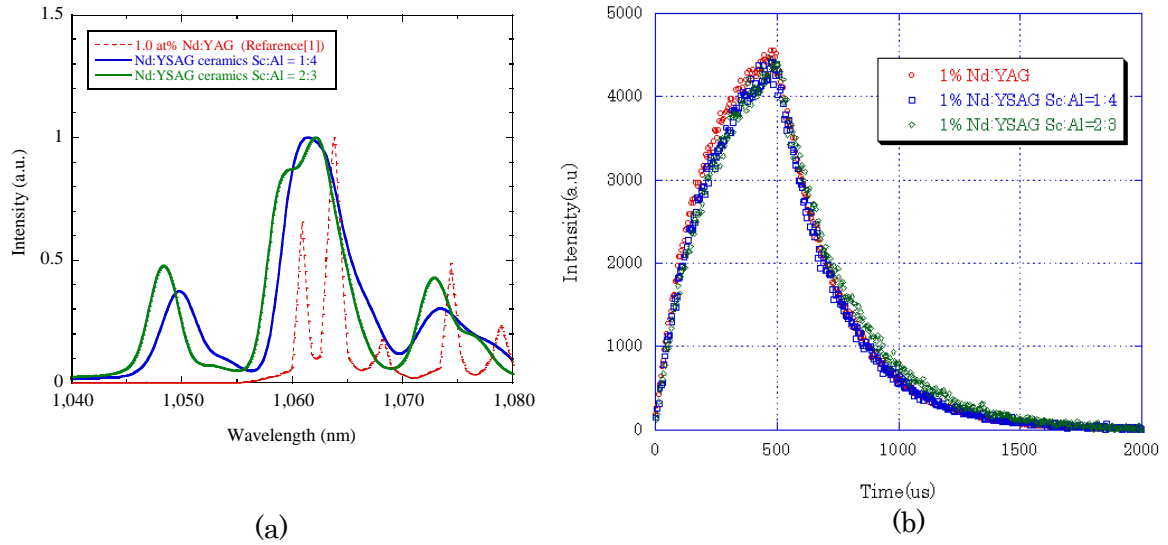


Figure 6-5(a) Emission spectrum and (b) fluorescence lifetime of the Nd:YAG ceramics.

## 7. Fabrication and evaluation of the multi-layered Nd:YAG composite

One of the challenge of this work is to make a complicated composite with layers of Nd:YAG, Nd:YAG (Sc:Al=1:4) and Nd:YAG (Sc:Al=2:3) in one-piece. By diffusion bonding technique, YAG composite (i.e.,  $\text{Nd:Y}_3\text{Al}_5\text{O}_{12}/\text{Nd:Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}/\text{Nd:Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$ ) was successfully fabricated for the first time. A series of thin slices (thickness up to 2mm) of each composition was prepared from the tablets of the above. Then the surfaces of thin slices to be bonded were finished by precision polishing (see figure 7-1). Thin slices with three different compositions were stacked in order as shown in figure 7-2(a), and it was heat treated at high temperature for diffusion bonding. The obtained composite sample was AR coated for laser oscillation test as seen in figure 7-2(b).

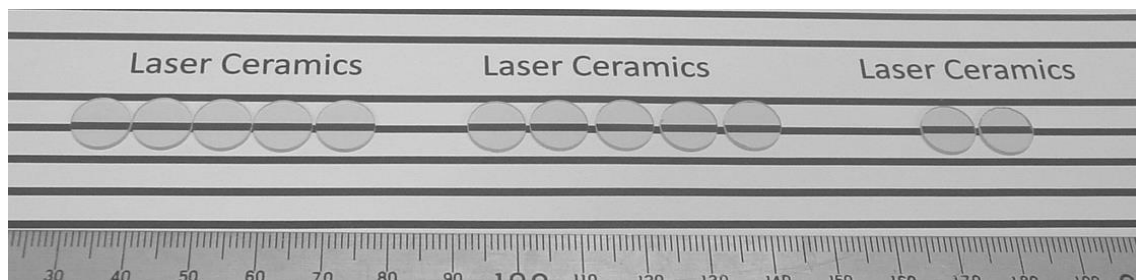


Figure 7-1 Thin slices of Nd:YAG ceramic samples with various compositions.

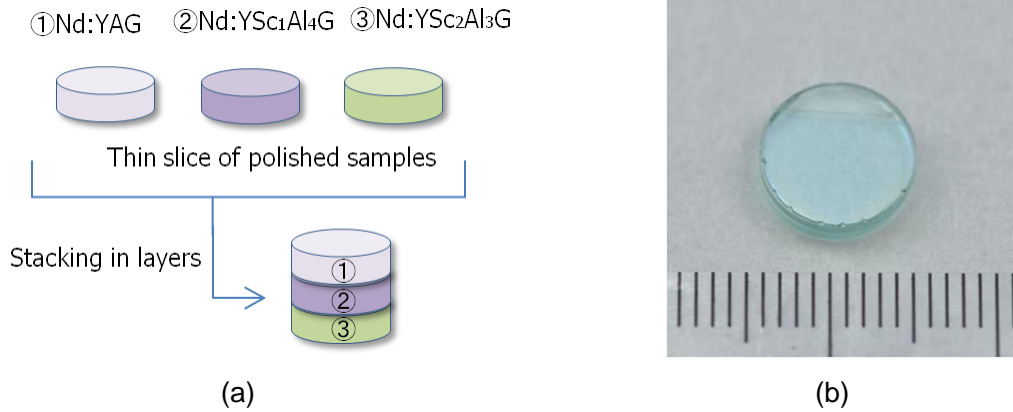


Figure 7-2(a) Schematic of bonding, and (b) external view of the obtained three layered Nd:YSAG composite after AR coating for laser oscillation test.

Regardless of the complicated compositions, it was confirmed that highly transparent Nd:YSAG ceramic composites can be fabricated with diffusion bonding method. The composite sample was observed under transmission polarizer. The results are shown in figure 7-3. No mechanical stress due to bonding was recognized. In addition, the optical quality near the bonding interfaces (i.e. Nd:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>/Nd:Y<sub>3</sub>Sc<sub>1</sub>Al<sub>4</sub>O<sub>12</sub> and Nd:Y<sub>3</sub>Sc<sub>1</sub>Al<sub>4</sub>O<sub>12</sub>/Nd:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> interfaces) was also inspected by transmission optical microscope and results are shown in figure 7-4(a) and (b). At these interfaces, no mechanical stress nor formation of secondary phases were observed. But as for Nd:Y<sub>3</sub>Sc<sub>1</sub>Al<sub>4</sub>O<sub>12</sub>/Nd:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> interface, some residual pores and localized defects were observed. It can be considered that the defects were caused not because of the bonding process but because of the quality of the Nd:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> ceramics that can be speculated from the microstructure shown in figure 5-3.

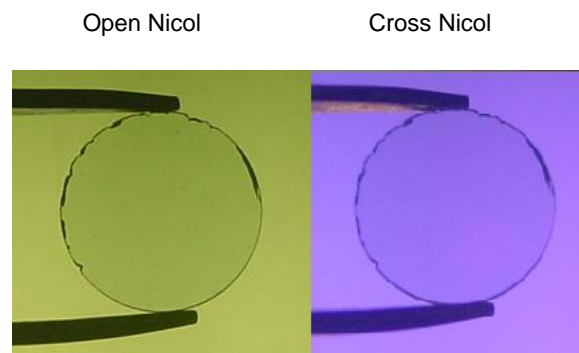


Figure 7-3 Observation under transmission polarizer.

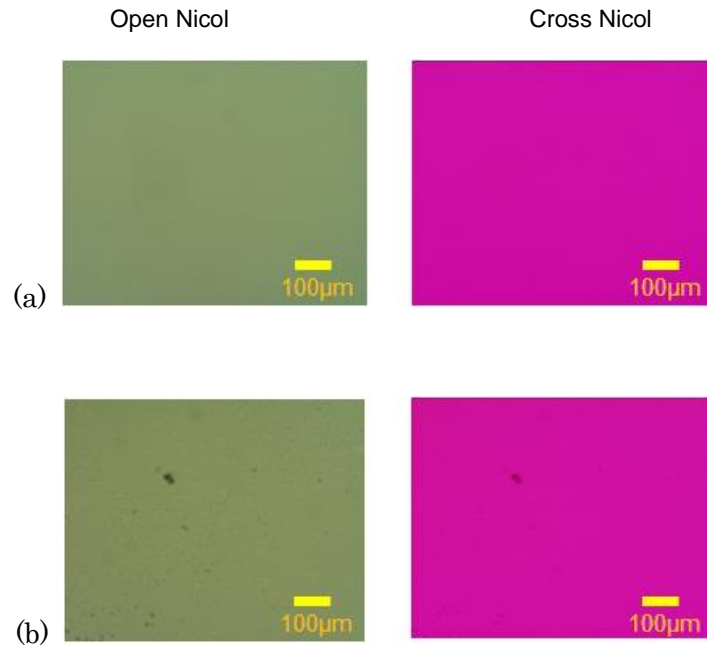


Figure 7-4 Optical quality near the interface of (a)  $\text{Nd:Y}_3\text{Al}_5\text{O}_{12}/\text{Nd:Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}$  and (b)  $\text{Nd:Y}_3\text{Sc}_1\text{Al}_4\text{O}_{12}/\text{Nd:Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$  observed under transmission optical microscope.

An evaluation system that can perform both emission spectrum measurement and also laser oscillation spectrum was constructed. Emission spectrum and laser emission spectrum were measured by pumping with 808nm. Emission spectrum of the Nd:YSAG ceramic composite samples was measured in the same way as described above. The results are shown in figure 7-5. Since the measurement was conducted at room temperature (300K), the spectra were not sharp. FWHM of the emission spectrum for the 1at%Nd:YAG which was used as a reference was 1.1 nm [1]. In contrast, the spectral width of the fabricated multi-layered Nd:YSAG composite was 6.8 nm and the center wavelength of the oscillation spectrum was 1063.5 nm. Compared to the reference 1at%Nd:YAG, it was confirmed that the bandwidth of the Nd:YSAG composite in FWHM was about 6 nm broader than that of the reference material and the center wavelength was also shifted about 0.6 nm toward shorter wavelength direction. It can be considered that it was due to the integral effect of Nd:YSAG (Sc:Al = 1:4) layer and Nd:YSAG (Sc:Al = 2:3) layer by forming composite structure.

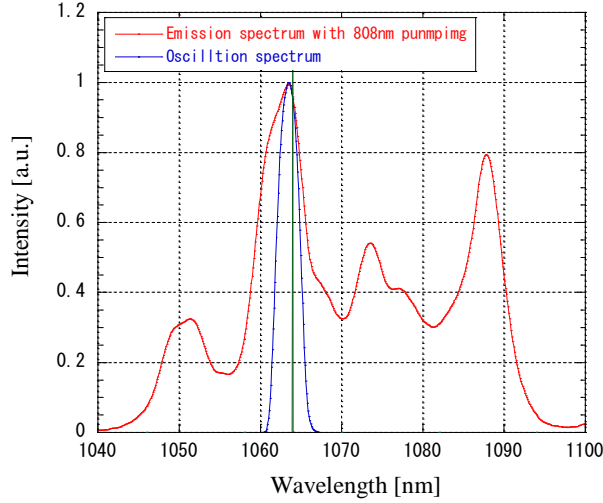


Figure 7-5 Emission spectrum with 808 nm pumping and laser oscillating spectrum for multi-layered Nd:YSAG composite.

Then, laser oscillation experiment was performed for the fabricated Nd:YSAG composite sample. A 1at% Nd:YAG ceramic was also prepared for reference. LD of 808nm was used as a pumping source. The sample was attached in a copper heat sink by using heat conduction grease. AR coating was done for all polished surface. A resonator was constituted by putting the composite sample between two flat mirrors. The mirror of 95% of transmittance was selected as an output coupler (O.C.). The resonator length was approximately 20mm. Relationship between absorbed power and output power is shown in figure 7-6. For the composite sample, as absorbed power increased, the laser output increased linearly up to 1 W. Slope efficiency about 38% was confirmed in this experiment. In contrast, output power of about 2 W was obtained from 1at% Nd:YAG ceramic sample. Slope efficiency about was 75%. These results revealed that the Nd:YSAG composite sample reached a laser grade although it is not yet a laser grade with high quality. It is necessary to improve the optical quality of the complicated composition of  $\text{Nd:Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$  ceramics as shown in figure 7-4(b) in order to improve the optical quality of the multilayered Nd:YSAG composite as a whole.

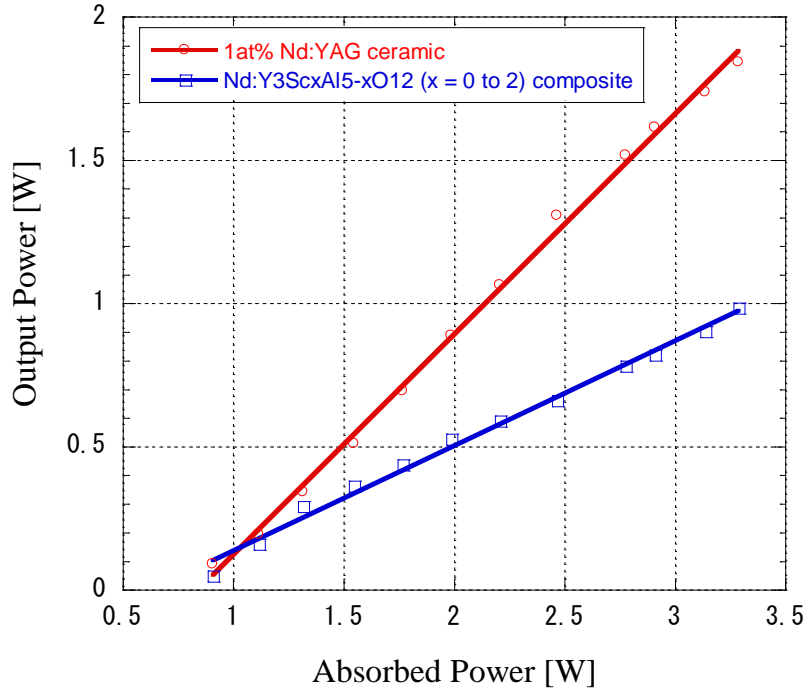


Figure 7-6 Relationship between absorbed power and output power for 1.0at% Nd:YAG ceramic and a three layered Nd:YSAG composite.

## 8. Conclusions

Advanced garnet ceramics with complicated compositions were successfully fabricated in CLNGG, CLNTGG and YSAG systems. All ceramic samples were highly densified and translucent quality with garnet single-phase. Broadband emissions were confirmed that cannot be observed in normal YAG materials. Nd doped  $\text{Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ( $x = 0$  to 2) ceramics is one of the candidates for high energy laser materials with broadband. It was confirmed that PL spectral shift and width can be controlled with a composition ratio of Sc to Al in the case of Nd doped  $\text{Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ( $x = 0$  to 2). When the Sc to Al ratio become larger, the fluorescence lifetime has become longer and spectral width has become larger than that of 1at%Nd:YAG. A composite structure with three different compositions in Nd doped  $\text{Y}_3\text{Sc}_x\text{Al}_{5-x}\text{O}_{12}$  ( $x = 0$  to 2) ceramics was successfully fabricated for the first time to obtain a broadband spectrum. The emission spectrum of YSAG composite was more than 5 times broader compared to that of 1at% Nd:YAG reference. Although  $\sigma$  (cross-section of stimulated emission) value was not measured in this work, if the  $\sigma$  value is larger than that of the conventional Nd:YAG, then it is certain that the product of  $\sigma$  (cross-section of stimulated emission) and  $\tau$  (life time of fluorescence) will be increased. Although the slope efficiency for the Nd:YSAG composite was about 38%, it was lower than that of the 1at%Nd:YAG (75%). It was confirmed that the Nd:YSAG composite sample reached a laser grade. But it is still necessary to improve the optical quality of the complicated composition of  $\text{Nd:Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$

ceramics in order to improve the optical quality of the multilayered Nd:YSAG composite as a whole. This kind of YSAG composite with complicated compositions is highly expected for the application in high energy laser field and short pulse laser. Besides, if a fabrication technology to produce CLNGG and CLNTGG types of ceramics with laser grade can be established, they will become one of the promising new laser materials. These results open the possibility of the creation of high-energy laser elements in addition to the glass medium for the first time and will lead laser system to various new applications.

### **Reference**

[1] Y. Sato, J. Saikawa, T. Taira and A. Ikesue. "Characteristics of Nd<sup>3+</sup>-doped Y<sub>3</sub>ScAl<sub>4</sub>O<sub>12</sub> ceramic laser". Optical Materials 29 (2007), 1277-1282.

### **Acknowledgement**

This work was supported by AFOSR/AOARD through funds received from Roppongi office, Tokyo AOARD Grant Number: FA2386-11-1-4007